

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of :)	
Jean-Marie BERNARD et al.)	Group Art Unit: 1762
)	
US Utility Patent Application No. 10/508,940)	Examiner: Rachel F. GORR
)	
Filed: September 27, 2004)	Confirmation No.: 5235
)	
For: LOW-VISCOSITY POLYISOCYANATE)	
COMPOSITION HAVING HIGH)	
FUNCTIONALITY AND METHOD FOR)	
PREPARING SAME)	

DECLARATION UNDER C.F.R. § 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Jean-Marie Bernard, declare as follows:

- 1) I was awarded a Doctor in physicochemistry of biologic and synthetic macromolecules from the Lille 1 university.
- 2) Currently, I am a Senior staff scientist in RHODIA in the Lyons Research Center, 85, rue des Frères Perret, BP 62, F-69192 Saint-Fons Cedex, France.
- 3) My Curriculum Vitae, Research Experience and list of publications are attached hereto as Appendix I.
- 4) US Patent 5,750,629 by Laas *et al.* discloses a process for the preparation of polyisocyanate mixtures, and more precisely of bis(isocyanato-alkyl)mono-uretdiones. In Example 1, at column 6, HDI is reacted with IPDI, TMPD and tri-n-octylphosphine. After termination of the reaction with MTS and removal of the diisocyanate mixture by distillation in a thin film evaporator, a product having a NCO content of 19.5% was obtained.
- 5) As shown in Appendix II, I made the calculations of the functionality of the mixture as prepared in Example 1 of the Laas patent, which is 2.56.

I HEREBY DECLARE that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful, false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful, false statements may jeopardize the validity of the application or any patent issued thereon.

Date :

the 05th of September 2006

Signature : Jean-Marie BERNARD

A handwritten signature in black ink, appearing to be 'J. Bernard', written over a set of horizontal lines.

APPENDIX I

Dr. BERNARD, Jean Marie

Born on December 23, 1954 in Somain, France

I studied chemistry and biochemistry in Lille University ("maîtrise").

In 1981, I obtained a diploma (PhD) in physicochemistry of biologic and synthetic macromolecules from the Lille 1 University.

From the 26th of January 1981 to the 1st of September 1986, I worked in the Research Center of CHOAY Institute (French Pharmaceutical Company) in Paris. During this time, I have been in charge of the synthesis of new active molecules for different therapeutic domains: immunochemistry, heparinotherapy, cardiovascular activity and cancerotherapy.

I have acquired an experience in organic chemistry and more specifically in lipid, glycan and peptide chemistry.

I am the author of patents and publications on these topics:

- Patents FR 2551758; FR 2564096; FR 2581069.

- Publications in:

J. Biol. Response Modif. (1987), 6(6), 678 – 691

Int. J. Pept. Prot. Research (1987), 29 (4) 455 – 463

J. Biol Response Modif. (1985), 4(5) 464 – 474

Peptides Proceedings European, Peptides symposium 18th (1984), 443- 446
Publisher Almqvist and Wiksell, Stockholm)

From the 1st of September 1986 to mars 1992, I worked in RHÔNE-POULENC as chemical engineer in peptide chemistry and I was in charge of a team (8 members) as technical leader. In mars 1992, I have acquired the function of research associate.

Since 1993, in the Research Center of Lyons (CRL) (RHODIA) and Research Industrialisation and Technology Center (CRIT) of Décines (RHODIA), I am in charge of research of new molecules and new process in the polyisocyanate field for coatings.

I am the author of more than 20 patents in the different fields: peptide chemistry, organic chemistry and isocyanate chemistry (organic, aqueous and powders compounds and their applications).

I am author of publications in Synlett (1993), 9, 680 – 682; Tetrahedron (1994), 50(2), 497–503; Tetrahedron Letters (1994), 35(47), 8783–8786; Tetrahedron Letters (1995), 36(8), 1267–1270.

I am co-author of the following books:

"The roots of Organic Development", published in 1996 by ELSEVIER (ISBN no. 0 444 824346 0)

"Waterborne and Solvent Based Surface Coating Resins and their Applications" published in 1998 by John WILEY and Sons (Chichester / New York / Weinheim / Brisbane / Toronto/ Singapore) in association with SITA Technology Ltd (London, UK). (ISBN no. 0471 978868)

I was awarded the RHÔNE-POULENC Research Price in 1991.

I am currently Senior Staff Scientist in the expert file of RHODIA.

- APPENDIX II. -

The composition disclosed in Example 1 of the Laas *et al.* patent (US 5,750,629) is obtained after removal by distillation of the HDI true dimer.
It should be kept in mind that the heavy oligomers comprise carbamate structures, since diol was introduced in the reaction mixture.

In said Example 1, the HDI transformation rate is 15%.
That is, for 10 kg of HDI, an amount of 1500 g ($10,000 \text{ g} \times 15\%$) is transformed into isocyanate oligomer.

To this amount must be added the diol (1,3-butanediol, 100 g; 1.1 moles), the catalyst (tri-n-butylphosphine, 30 g) and the terminating agent (methyl toluenesulphonate, 28 g), that is a **total amount of polyisocyanate mixture** of $(1500 + 100 + 30 + 28 =)$ 1658 g.

Laas *et al.* disclose that (see column 6, lines 41-43), after distillation of the remaining monomer, the content of oligomer mixture is as follows:

- 39.0% of HDI uretdione, i.e. a weight amount of:
 $(1658 \times 39\% =)$ **646.62 g of true dimer;**
- 22.6% of HDI isocyanurate, i.e. a weight amount of:
 $(1658 \times 22.6\% =)$ **374.71 g of trimer; and**
- 38.1% of higher oligomers, i.e. a weight amount of:
 $(1658 \times 38.1\% =)$ **631.7 g of higher oligomers.**

Moreover, 100 g (1.1 moles) of diol were involved in the reaction, i.e. a weak amount as compared to HDI (59.5 moles).

It may then be concluded that the totality of the diol reacted with HDI (2 moles of HDI react with 1 mole of diol).

Consequently, the resulting **urethane oligomer** (neither containing isocyanurate nor dimer) is in an amount of **469.6 g**, calculated as follows:

$100 \text{ g diol (1.1 moles)} + (2.2 \text{ moles HDI} \times 168 \text{ (molecular weight of HDI)}) = 100 \text{ g} + 369.6 \text{ g} = 469.6 \text{ g of polyurethane.}$ This amount represents $(100 \times 469.6 \text{ g} / 631.7 \text{ g} =)$ **74.3% of the higher oligomers.**

The amount of **heavy non urethane oligomer** compounds possibly comprising more than one trimer ring or more than one dimer ring or possibly comprising at least one trimer ring and one dimer ring is calculated as being: $631.7 \text{ g} - 469.6 \text{ g} = 162.1 \text{ g}.$

This represents a percentage to the initial polyisocyanate mixture of $(100 \times 162.1 / 1658 =)$ 9.8%, that is to say an amount of $(100 \times 9.8 / 38.1 =)$ **25.7% of the higher oligomers.**

After removal of the pure monouretdione diisocyanate from 1000 g of initial polyisocyanate mixture, 670 g of new liquid polyisocyanate mixture is obtained (see column 6, line 48). The oligomer composition given by Laas *and al.*, column 6, lines 55-57 is the following:

- HDI uretdione (true dimer): 19.4% ;
- HDI isocyanurate (true trimer): 28.7% ; and
- and higher oligomers: 51.9%.

The composition of the distillate (pure monouretdione diisocyanate) given by Laas *et al.*, at column 7, lines 4-6, shows there is practically no higher oligomers (0.6%). The ratio between urethane oligomers and heavy non urethane oligomers in the new liquid polyisocyanate mixture (obtained after separation of dimer) may be considered as practically the same as the initial polyisocyanate mixture (obtained after removal of untreated excess HDI monomer).

Consequently, the amount of **urethane oligomer** (neither containing trimer nor dimer) in the new liquid polyisocyanate mixture represents $(51.9 \times 74.3\% =) 38.56\%$, and the amount of **heavy non urethane oligomer compounds** possibly comprising more than one trimer ring or more than one dimer ring or possibly comprising at least one trimer ring and one dimer ring represents $(51.9 \times 25.7\% =) 13.34\%$.

Therefore, the final composition of liquid polyisocyanate is as follows:

- HDI uretdione (true dimer): 19.4% (given by Laas *et al.*);
- HDI isocyanurate (true trimer): 28.7% (given by Laas *et al.*);
- Urethane oligomer: 38.56% (calculated as above); and
- Heavy non urethane oligomers compounds: 13.34% (calculated as above).

Note that the amount of **heavy non urethane oligomers** is much lower than the one claimed in our application, which is at least 40% by mass (see c) in Claim 1).

The functionality of the mixture may be calculated (as explained on pages 15-16 of the present application) as the sum of the functionalities of each of the components of the mixture. The functionality, calculated for 100 g. of the **new liquid polyisocyanate mixture** is as follows:

- i) Functionality of the dimer $(2) \times \% \text{ of dimer } (19.4\%)$ (Laas data),
- ii) Functionality of the trimer $(3) \times \% \text{ of trimer } (28.7\%)$ (Laas data),
- iii) Functionality of the oligourethane $(2) \times \% \text{ of oligourethane } (38.56\%)$ (see above),
- iv) Functionality of the heavy non urethane oligomer compounds $(4^*) \times \% \text{ of the heavy non urethane oligomers } 13.34\%)$ (see above).

The sum of i) to iv), is:

$$(19.4 \times 2 / 100) + (28.7 \times 3 / 100) + (38.56 \times 2 / 100) + (13.34 \times 4 / 100) = 0.39 + 0.86 + 0.77 + 0.54 = 2.56.$$

The functionality of the polyisocyanate mixture disclosed by Laas *et al.* in Example 1 is 2.56, that is to say less than 3.

*. A high value (4) for the functionality of heavy non urethane oligomers is used here. This functionality is overestimated, considering the presence of higher non urethane oligomers comprising more than one dimer function which have a functionality as low as 2.